(12)

(19) Japan Patent Office (JP)

(12) Japanese Patent Laid-Open Application Publication (A)

(11) Patent Laid-Open Application No. Hei 8-109178

(43) Date of publication of application: April 30, 1996

(21) Patent Application: Patent Appln. No. Hei 6-270211

(22) Filing Date: October 11, 1994

(54) [Title of the Invention] METHOD OF MANUFACTURING LOW-CAFFEINE TEA POLYPHENOL

(57) [Abstract]

[Constitution]

A method of manufacturing low-caffeine tea polyphenol characterized by dissolving or suspending a tea extract in water or a water-containing organic solvent and bringing this solution or suspension into contact with a synthetic adsorbent under an alkaline condition to adsorb and remove caffeine.

[Advantages]

According to the present invention, tea polyphenol of which caffeine content is low can be efficiently manufactured by a simple and safe method.

[Claims for the Patent]

[Claim 1]

A method of manufacturing low-caffeine tea polyphenol characterized by dissolving or suspending a tea extract in water or a water-containing organic solvent and bringing this solution or suspension into contact with a synthetic adsorbent under an alkaline condition to adsorb and remove caffeine.

[Claim 2]

The method of manufacturing the tea polyphenol according to claim 1, wherein the tea extract is a hot-water extract of tea, an organic-solvent extract of tea, or a product obtained by treating the hot-water or organic-solvent extract with an organic solvent, a membrane, a resin, or an adsorbent.

[Claim 3]

The method of manufacturing the tea polyphenol according to claim 1, wherein the synthetic adsorbent has the main component of any one of styrene, styrene divinylbenzene, acrylic, methacrylic, acrylic acid ester, amide, dextran, cellulose, and polyvinyl.

[Claim 4]

The method of manufacturing the tea polyphenol according to claim 1, wherein the alkaline condition is a pH of 7 to 14.

[Detailed Description of the Invention]

[0001]

[Industrial Application Field]

The present invention relates to a method of manufacturing tea polyphenol, and more specifically relates to a method of manufacturing low-caffeine tea polyphenol.

[0002]

[Conventional Art and Problems to be Solved by the Invention]

Tea polyphenol has an anti-oxidative activity (Japanese Patent Laid-Open Nos. 59-219384 and 1-268683), an anti-bacterial/bacteriostatic activity (Japanese Patent Laid-Open Nos. 2-276562 and 3-246227), and an antitoxic activity (Japanese Patent Laid-Open Nos. 2-304079 and 2-306915), and also activities of controlling biological functions, such as an activity of suppressing an increase in cholesterol (Japanese Patent Laid-Open No. 60-156614), an activity of suppressing an increase in blood pressure (Japanese Patent Laid-Open No. 63-214183), and an activity of suppressing an increase in blood sugar (Japanese Patent Laid-Open No. 4-253918). Accordingly, tea polyphenol is expected to be used in various fields such as drugs and agricultural chemicals, in addition to foods.

[0003]

Tea polyphenol is manufactured by various methods, and in general, is extracted from tea leaves with hot water or an organic solvent, in many cases. In such cases, a large amount of caffeine is unavoidably contained in the tea extract. Caffeine has physiological activities such as a central nerve-excitatory activity, a cardiotonic activity, and a diuretic activity, and is thereby widely used in drugs such as medicines for headache or cold. However, the strong physiological activities of caffeine cause dizziness, insomnia, palpitation, nausea, and so on depending on the dose or individual variation. Accordingly, the caffeine contents in foods and drinks are an important matter for people suffering caffeine allergy. Furthermore,

caffeine is suspected to increase the blood cholesterol, which causes arteriosclerosis or myocardial infarction, in addition to the aforementioned acute intoxication and is under investigation at present (Kato and Yoshida, (1981) Nutr. Rep. Inter., 23: 825). Furthermore, there is reported that the intake of caffeine increases the excretion of calcium to cause hypercalciuria (Heaney, R.P. and Recker, R.R., (1982) J. Lab. Clin. Med., 99: 46). From such reasons, tea extracts are also required to contain a smaller amount of caffeine.

[0004]

Conventionally, in typical methods, caffeine is removed, for example, by extraction using a chlorine solvent (Japanese Patent Publication Nos. 2-22755 and 2-12474), by extraction using supercritical carbon dioxide (Japanese Patent Laid-Open Nos. 48-4692 and 1-289448), by adsorption with activated carbon or the like (Japanese Patent Publication No. 1-45345), or by extraction using an acid aqueous solution (Japanese Patent Application No. 5-344744).

[0005]

However, these methods have the respective problems. The extraction using a chlorine solvent has problems of safety and residue of the solvent containing chlorine and is also environmentally unpreferable. The extraction using supercritical carbon dioxide requires large-scale facilities and is hence expensive in the initial cost and is also low in productivity. The adsorption with activated carbon or the like has disadvantages that tea polyphenol is also adsorbed together with caffeine that should be removed, and thereby the loss of tea

polyphenol is large. The extraction using an acid aqueous solution requires an organic solvent such as ethyl acetate and also has a low recovery rate of tea polyphenol.

[0006]

[Means for Solving the Problems]

The present inventors have conducted intensive studies in order to solve the aforementioned problems, and as a result, have found a method that can reduce the amount of caffeine in a tea extract by simple, efficient, and safe means. Thus, the present invention has been accomplished.

[0007]

That is, the present invention relates to a method of manufacturing low-caffeine tea polyphenol characterized by dissolving or suspending a tea extract in water or a water-containing organic solvent and bringing this solution or suspension into contact with a synthetic adsorbent under an alkaline condition to adsorb and remove caffeine.

[0008]

Tea used in the present invention may be any type without regard to whether or not the tea is fermented or not, and examples of the tea include green tea, black tea, oolong tea, and pu-erh tea. The tea extract obtained by extracting tea is, for example, a hot-water extract or an organic-solvent extract of tea obtained by a usual method, or a product obtained by treating the hot-water or organic-solvent extract with any of various types of organic solvents, membranes, resins, or adsorbents. The caffeine content in each of these extracts is usually about 5 to 15%. Here, the extraction of tea with hot

water is a method conducted by immersing tea leaves for extraction in hot water or boiling water of a weight several times that of the tea leaves. The extraction of tea with an organic solvent is a method conducted by immersing tea leaves in an organic solvent of a weight several times that of the tea leaves, and examples of the organic solvent include tea polyphenol-dissolving organic solvents, such as acetonitrile, methanol, ethanol, ethyl acetate, acetone, methyl ethyl ketone, methyl isobutyl ketone, tetrahydrofuran, and dioxane; water-containing solvents thereof; and solvent mixtures thereof.
[0009]

The organic solvent-treated product is an extract obtained by further re-extracting the aforementioned hot-water extract or the organic-solvent extract with the aforementioned organic solvent or by removing components other than tea polyphenol from the aforementioned hot-water extract or the organic-solvent extract by extraction with an organic solvent such as chloroform, dichloromethane, or hexane. The membrane-treated product is a product obtained by membrane filtration or dialysis treatment of the hot-water extract or the organic-solvent extract. The product obtained by treatment with a resin or an adsorbent is a product obtained by bringing the aforementioned hot-water extract or the organic-solvent extract into contact with a synthetic adsorbent, activated carbon, or the like for collecting tea polyphenol by attaching and detaching or removing components other than tea polyphenol by adsorbing.
[0010]

In the present invention, the aforementioned tea extract is dissolved or suspended in water or a tea polyphenol-dissolving water-containing organic solvent, such as ethanol, methanol, acetone, tetrahydrofuran, or dioxane, or a solvent mixture thereof; and then alkalifying the resulting solution or suspension and bringing it into contact with a synthetic adsorbent. Here, the water-containing organic solvent may be any type and is preferably ethanol or methanol and particularly preferably ethanol. Furthermore, the concentration of the organic solvent is preferably 0 to 50% (v/v) and more preferably 0 to 30% (v/v). The synthetic adsorbent may be any type, and examples thereof include synthetic adsorbents having the main components of styrene, such as XAD-16 (Rohm and Haas); styrene divinylbenzene, such as SEPABEADS HP21 (Mitsubishi Chemical Industries); acryl, such as DIAION WK20 (Mitsubishi Chemical Industries); metacryl, such as SEPABEADS HP2MG (Mitsubishi Chemical Industries); acrylic acid ester, such as XAD-7 (Rohm and Haas); amide, such as XAD-11 (Rohm and Haas); dextran, such as SEPHADEX LH-21 (Pharmacia); cellulose, such as INDION DS-3 (Phenix Chemicals); or polyvinyl, such as SEPABEADS FP-HG (Mitsubishi Chemical Industries). An alkalinity of pH 7 to 14 is effective, and pH 9 to 11 is optimum. The contact may be conducted by any method such as a batch system or a column system.

[0011]

With this treatment, a tea polyphenol solution containing a smaller amount of caffeine can be obtained. That is, the caffeine content of the tea polyphenol solution is about 0.1 to

1.0% of the solid content. The tea polyphenol solution containing a smaller amount of caffeine can be used directly or after neutralization with acid or after desalination, concentration, and drying by a usual method. In addition, the tea polyphenol solution can be used as a raw material for manufacturing tea polyphenol of high purity.

[0012]

The low-caffeine-containing tea polyphenol of the present invention does not practically contain caffeine. Accordingly, there is no concern for the above-mentioned negative effects of caffeine, and the tea polyphenol can be effectively used as a health promoting food, a health maintenance food, a health restoring food, or the like. These foods are imparted with an essential activity of polyphenol, for example, physiological activities such as a cholesterol increase-suppressing activity and an in vivo antioxidative activity.

[0013]

The low-caffeine-containing tea polyphenol of the present invention can be applied to the fields of, for example, foods such as seasonings, Japanese sweets, cakes, ices, syrups, processed fruits, processed vegetables, pickles, animal meat products, fish products, dainties, canned or bottled products, alcoholic liquors, soft drinks, and instant foods and drinks; and various solid, paste, or liquid favorite products, cosmetics, or drugs such as cigarettes, tooth pastes, lipsticks, lip balms, internal medicines, troches, liver oil drops, mouth deodorants, cachous, and gargles.

[0014]

[Examples]

The present invention will now be described in more detail with reference to Examples, but is not limited thereto in any way.

Example 1

Ten grams of a green tea extract (containing 7% of caffeine and 30% of catechin) was dissolved in 20 mL of water, and the resulting solution was applied to a glass column (40 mm I.D. × 300 mm) packed with 300 mL of a synthetic adsorbent, SP-207 (Mitsubishi Chemical Industries). Then, 1500 mL of a buffer solution at pH 10 was applied to the column (SV = 2), and a fraction of tea polyphenol was collected. This fraction was subjected to desalination, concentration, and drying to give 2.9 g of a powder.

[0015]

The thus obtained sample was analyzed by high-performance liquid chromatography to confirm that the caffeine content was 0.2% and the catechin content was 64%. The analysis conditions are shown below. Figure 1 shows chromatograms of the sample before and after the treatment of removing caffeine.

[0016]

High-performance liquid chromatography analysis conditions

Column: Shiseido CAPCELL PAK AG-120 S-5 ODS 4.6 mm I.D. × 250 mm

Eluant: acetonitrile : ethyl acetate : 0.05% phosphoric acid aqueous solution = 12:2:86

Flow rate: 1 mL/min

Detector: ultraviolet absorptiometer 280 nm

Temperature: 40°C

[0017]

Example 2

Twenty grams of a green tea extract (containing 7% of caffeine and 30% of catechin) was dissolved in 100 mL of water, and the resulting solution was applied to a glass column (40 mm I.D. × 300 mm) packed with 300 mL of a synthetic adsorbent, HP-20 (Mitsubishi Chemical Industries). Then, 1500 mL of a solution consisting of a buffer solution at pH 11 and ethanol at a ratio of 4:1 (v/v) was applied to the column (SV = 2), and a fraction of tea polyphenol was collected. This fraction was subjected to desalination, concentration, and drying to give 8.3 g of a powder. The thus obtained sample was analyzed by a method as in Example 1 to give the results that the caffeine content was 0.6% and the catechin content was 56%. Figure 2 shows chromatograms of the sample before and after the treatment of removing caffeine.

[0018]

Example 3

Fifteen grams of a green tea extract with ethyl acetate (containing 11% of caffeine and 66% of catechin) was dissolved in 30 mL of a 20% ethanol solution, and the resulting solution was applied to a glass column (40 mm I.D. × 300 mm) packed with 300 mL of a synthetic adsorbent, HP-2MG (Mitsubishi Chemical Industries). Then, elution was conducted using the same solvent as in Example 2 to give 11.6 g of a powder. The thus obtained sample was analyzed by a method as in Example 1 to give the results that the caffeine content was 0.7% and the catechin

content was 73%. Figure 3 shows chromatograms of the sample before and after the treatment of removing caffeine.

[0019]

[Advantages of the Invention]

According to the present invention, tea polyphenol containing a smaller amount of caffeine can be efficiently manufactured by a simple and safe method.

[Brief Description of the Drawings]

[Figure 1]

Figure 1 shows the analysis results of Example 1, and the upper column shows the chromatogram of a sample before the treatment of removing caffeine, and the lower column shows the chromatogram of the sample after the treatment.

[Figure 2]

Figure 2 shows the analysis results of Example 2, and the upper column shows the chromatogram of a sample before the treatment of removing caffeine, and the lower column shows the chromatogram of the sample after the treatment.

[Figure 3]

Figure 3 shows the analysis results of Example 3, and the upper column shows the chromatogram of a sample before the treatment of removing caffeine, and the lower column shows the chromatogram of the sample after the treatment.

[Description of Symbols]

Each of peaks a, b, c, d, and e shows tea catechin.

5

mm）に充填した合成吸着剤HP－2MG（三菱化成（株）製）300mlに通液した。これを実施例2と同様の溶媒で溶出し、粉末11．6gを得た。このようにして得られたサンプルを実施例1と同様の方法で分析した結果、カフェイン含率0．7%、カテキン含率73%であった。また、図3に脱カフェイン処理前と処理後のサンプルのクロマトグラムを示す。

【0019】

【発明の効果】本発明によれば、簡便、且つ安全な方法でカフェイン含有量の少ない茶ポリフェノールを効率よく製造することができる。

【図面の簡単な説明】

6

【図1】　実施例1の分析結果であり、上段は脱カフェイン処理前のサンプルのクロマトグラムを示し、下段は処理後のサンプルのクロマトグラムを示す。

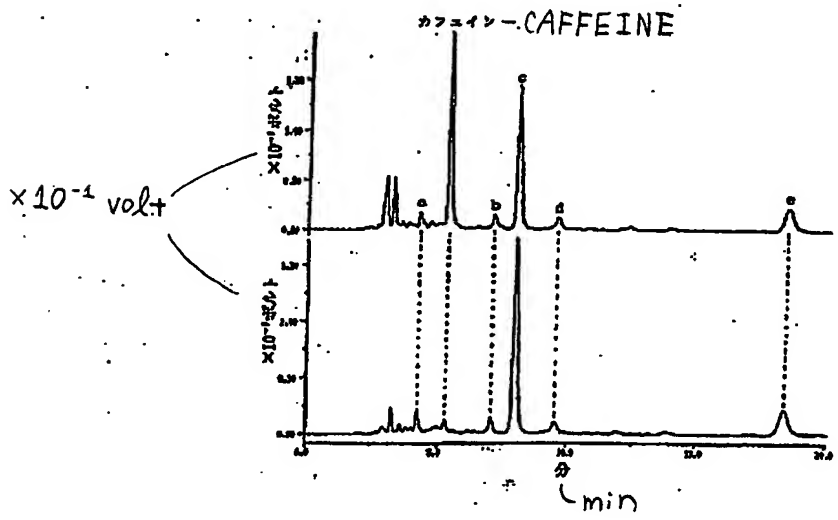
【図2】　実施例2の分析結果であり、上段は脱カフェイン処理前のサンプルのクロマトグラムを示し、下段は処理後のサンプルのクロマトグラムを示す。

【図3】　実施例3の分析結果であり、上段は脱カフェイン処理前のサンプルのクロマトグラムを示し、下段は処理後のサンプルのクロマトグラムを示す。

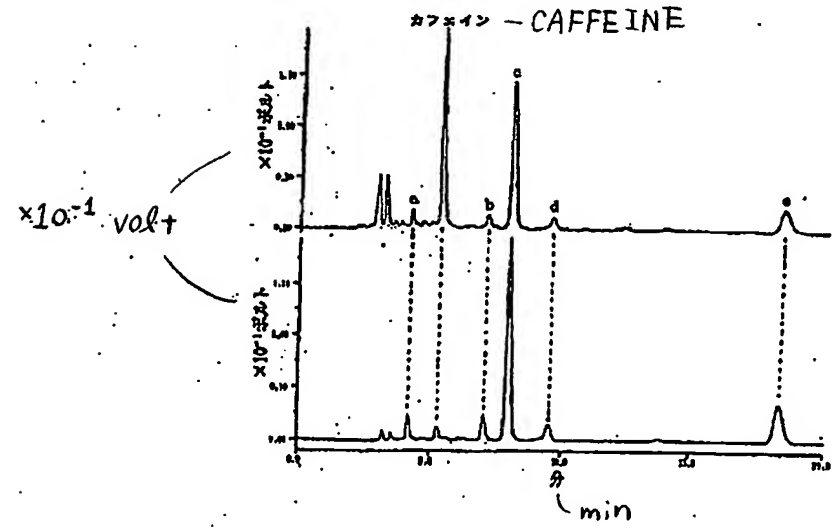
【符号の説明】

ピークa、b、c、d及びeはいずれも茶カテキンを示す。

【図1】



【図2】



【図3】